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The Role of Grain Boundary Energy on Grain Boundary Complexion Transitions

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The Role of Grain Boundary Energy on Grain Boundary Complexion Transitions

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Abstract

Grain boundary complexions are distinct equilibrium structures and compositions of a grain boundary and complexion transformations are transition from a metastable to an equilibrium complexion at a specific thermodynamic and geometric conditions. Previous work indicates that, in the case of doped alumina, a complexion transition that increased the mobility of transformed boundaries and resulted in abnormal grain growth also caused a decrease in the mean relative grain boundary energy as well as an increase in the anisotropy of the grain boundary character distribution (GBCD). The current work will investigate the hypothesis that the rates of complexion transitions that result in abnormal grain growth (AGG) depend on grain boundary character and energy. Furthermore, the current work expands upon this understanding and tests the hypothesis that it is possible to control when and where a complexion transition occurs by controlling the local grain boundary energy distribution.

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1. PROJECT SUMMARY

A grain boundary complexion is a distinct equilibrium structure and composition of a grain boundary. A complexion transformation is then a transition from a metastable to an equilibrium complexion at a specific temperature, pressure, chemical potential, and grain boundary character. Previous work indicates that, in the case of doped alumina, a complexion transition that increased the mobility of transformed boundaries and resulted in abnormal grain growth also caused a decrease in the mean relative grain boundary energy as well as an increase in the anisotropy of the grain boundary character distribution (GBCD). The current work will investigate the hypothesis that the rates of complexion transitions that result in abnormal grain growth (AGG) depend on grain boundary character and energy. Furthermore, the current work expands upon this understanding and tests the hypothesis that it is possible to control when and where a complexion transition occurs by controlling the local grain boundary energy distribution.

In the first experiment, grain boundary character and energy distributions were measured for Ca-doped yttria, which displayed abnormal grain growth as a result of a complexion transition that involved a Ca-rich intergranular film. It was found that grain boundary character anisotropy increased with the occurrence of abnormal grain growth – this was in agreement with previous studies in aluminas. Additionally, the relative grain boundary energy of the boundaries surrounding abnormal grains as well as the boundaries surrounding normal grains that were immediately adjacent to the abnormal grains were both 33% lower in energy in comparison to the boundaries of the untransformed normal grains that were further away from the abnormal grains. These results gave insight into the propagation of an intergranular film complexion transition beyond just the boundaries surrounding abnormal grains, and through triple junctions to boundaries surrounding normal grains that are in close proximity to the abnormal grains.

The second experiment investigated the effects of yttrium and lanthanum doping on the grain boundary character distribution of alumina. It was found that La doping resulted in a random misorientation angle distribution and a strong preference for (0001) grain boundary planes. The grain boundary character distributions at each temperature were self-similar, indicating that there was no complexion transition in La-doped alumina sintered between 1500 °C and 1600 °C. Y doping resulted in a bimodal grain size distribution at 1500 °C where there was an increasing population of $60^\circ/[0001]$ grain boundaries with temperature and the number of abnormal grains. Additionally, the population of $\{01\bar{1}2\}$ planes increased from the unimodal grain size sample sintered at 1450 °C to the sample exhibiting a bimodal grain size distribution that was sintered at 1500 °C. The microstructure at 1600 °C had a greater population of $\{11\bar{2}0\}$ planes and there was a minimum population of (0001) planes. The Y and La co dopants resulted in a self-similar grain boundary character distribution that exhibited local maxima of both (0001) and $\{01\bar{1}2\}$ planes and $60^\circ/[0001]$ grain boundaries. These results were the first to show an increase in grain boundary character anisotropy with abnormal grain growth and a further change in the preferred grain boundary plane with abnormal grain impingement. Additionally, this was the first time that a grain boundary misorientation has been shown to increase with the occurrence of abnormal grain growth.

The third experiment attempted to control complexion transitions by locally controlling the interfacial energy. $(11\bar{2}0)$ and (0001) single crystals of sapphire were embedded in Y-doped alumina and heated to 1500 °C for 8 hours. The (0001) interface was higher in energy than the $(11\bar{2}0)$ interface in the sample that was spark plasma sintered at 1300 °C. After the sample was heated to 1500 °C for 8 hours, a bimodal grain size distribution occurred. More abnormal grains grew along the high energy (0001) interface than along the $(11\bar{2}0)$ interface, and the energy of both interfaces decreased by 19 % and 27 % respectively. These results indicate that there is an energy dependence of complexion transitions, where high-energy interfaces require less thermal input to overcome the nucleation barrier to initiate a complexion transition while low energy interfaces need to be heated at longer times or at higher temperatures to nucleate the transition.

The fourth experiment analyzed the temperature and grain boundary character dependence of the rate of the complexion transformation in Y doped alumina and attempted to compare it to that of a bulk phase transformation. At most annealing temperatures and times, the higher energy (0001) interface was in contact with a larger area fraction of abnormal grains, while there were fewer large grains along the $(11\bar{2}0)$ interface. The rate of a complexion nucleation along each interface was greater for the higher energy (0001) interface at 1500 °C (0.69%/μm-hr) compared to the lower energy $(11\bar{2}0)$ interfaces (0.16%/μm-hr). At 1600 °C the nucleation rate was approximately the same (0.3%/μm-hr) however the higher energy (0001) interface exhibited a much higher initial percent of the interface that was transformed. Finally, there was no apparent field effect with the spark plasma sintering method. These results showed that there is a character and temperature dependence on the rate of nucleating complexion transitions. A maximum number of grain boundaries are metastable at any given temperature, and that number increases with temperature. At any temperature within the range of critical nucleation temperatures, the number of boundaries that transforms increases with time such that the highest energy boundaries transform at the shortest times.

In the fifth and final experiment, the grain boundary energy was measured as a function of temperature and composition in the Y-doped alumina system. It was found that in the 100 ppm Y-doped alumina experiment, the grain boundary energy increased with increasing temperature until 1450 °C. The energy then decreased by 20 % with the occurrence of high mobility grain boundaries. After 1550 °C the lower energy grain boundaries again increased with increasing temperature. The 500 ppm sample exhibited a decrease in grain boundary energy from 1350 °C to 1400 °C, indicating a shift from dilute to supersaturated grain boundaries. The energy then increased until 1450 °C, where at 1450 °C a second phase began to precipitate. Between 1450 °C and 1550 °C the mean energy decreased and both YAG precipitation and high mobility grain boundaries were observed. The total decrease in energy was 25 %. After 1550 °C, the energy increased in a similar manner as the 100 ppm sample. These results show that the grain boundary energy increases with the precipitation of a second phase and decreases with the occurrence of a complexion transition. It also indicated that the increase in grain boundary energy due to solute desorption with temperature could provide a driving force for a grain boundary complexion transition in the Y-doped alumina system.

Overall these investigations show that the grain boundary energy anisotropy and temperature are the important parameters influencing the nucleation of the grain boundary complexion transition in Y-doped alumina. With this knowledge, it is possible to control and predict when and where a

transition will occur. It is expected that similar trends can be observed in other ceramic and metallic systems. Specifically, it would be interesting to compare these results to a material that displays highly faceted abnormal grain morphology and complexion transitions involving intergranular films, such as the Ca-doped alumina system.

The details of these results are available in the publications listed in the References section of this report.

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